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## SHORT COMMUNICATION



# Corrosion of Gold in Biological Fluid - Urine

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Urinary catheters are the leading cause of nosocomical urinary tract infections (1). Moreover, the urinary tract infections are the most common predisposing factor for fatal gram-negative sepsis that originate in hospitals. Unfortunately, even widespread use of antibodies to cure the infection has not been successful (2). Thus, with all of the existing catheter and drainage bag designs, there is still a major problem with catheter-related infection (2). The application of electrochemical technology to control microbiological growth has been described in the patent literature for many decades. The long term effectiveness of gold electrodes during electrolysis in killing bacteria in heart fusion broth, and in synthetic urine with or without soy broth has been recently demonstrated by us (3,4). Based on this work, a research and development work is in progress to develop a commercially viable catheter device to prevent catheter - related urinary tract infections. Hitherto, the majority of the work on the corrosion of the gold (for example see ref (5)) is concern to the electronic industry. Consequently, a corrosion study of gold in synthetic urine was undertaken.

A custom built microampere generator was built to provide constant current (1 - 450 μA) or constant voltage (0.01 - 12.50 V). All experiments were conducted in synthetic urine solutions (Table 1) with or without 5% trypticase soy broth (TSB) (Table 2). The solutions were prepared using reagent grade chemicals as received, sterilized (filtration), and refrigerated prior to use. The solution pH was about 5.8. About 4 cms long gold wire (4N pure and 0.02 cm diameter, Johnson Matthey Chemicals Ltd.) was soldered to a copper tubing holder which provided a quick - fit connection between the wire electrode and the male connector of the constant current generator. About 1 cm of the wire electrode from the solder - joint was covered by a heat - shrinkable Teflon

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Table 1. Composition of Synthetic Urine (6)

| Compound | g/liter |
|---|---|
| $CaCl_2.2H_2O$ | 0.65 |
| $MgCl_2.6H_2O$ | 0.651 |
| NaCl | 4.6 |
| $Na_2SO_4$ | 2.3 |
| $Na_3$ citrate.$2H_2O$ | 0.65 |
| $Na_2$oxalate | 0.020 |
| $KH_2PO_4$ | 2.8 |
| KCl | 1.6 |
| $NH_4Cl$ | 1.0 |
| Urea | 25.0 |
| Creatinine | 1.1 |
| pH = 5.7 - 5.8 | |

Table 2. Composition of Trypticase Soy Broth

| Compound | |
|---|---|
| Trypticase Peptone | 17.0g |
| Phytone Peptone | 3.0g |
| NaCl | 5.0g |
| $K_2HPO_4$ | 2.5g |
| $H_2O$ | 1.0 liter |
| pH = 7.3 $\pm$ 0.2 | |

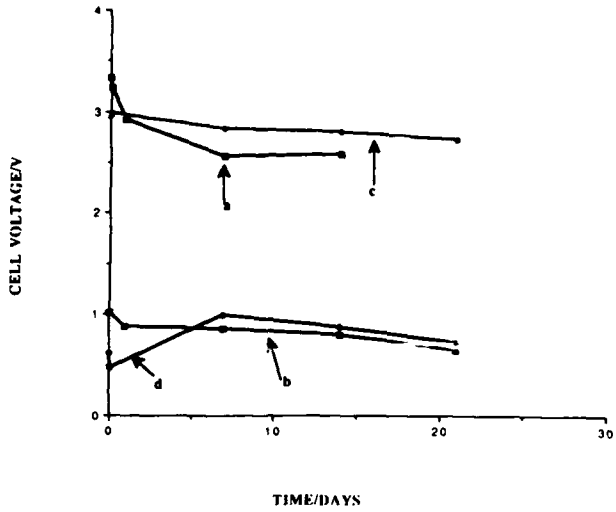
tubing. The Teflon tubing, solder - joint and 1 - 2 cms of copper tubing from the solder - joint were further covered by a heat - shrinkable polyolefin tubing. Finally, the uncovered electrode length was cut to 2 cms so that the solution exposed area of gold wire was 0.13 $cm^2$. A pair of gold electrodes, which were treated just before the experiment by dipping in 70% ethanol for 60 seconds and successfully drying at 200°C, were inserted in the vented, stoppered cell containing 10 ml of urine sample. The detailed description of the cell, instrumentation, and general experimental procedure have been described earlier (3,4). To simulate the catheter design condition, corrosion studies were carried out in a two-electrode configuration at room temperature and at various set currents. Cell voltages were monitored for 3 weeks or shorter depending on the electrode failure. Electrodes were weighed before and after the experiment on a Metler H51AR semimicrobalance with a precision of $\pm$ 10 $\mu$g.

The data for the gold dissolution in synthetic urine are given in Table 3. This is an average of 3 - 4 runs at each current. The cell voltage (Figure 1 (a and b)), in general, decreased with time of electrolysis to reach a steady state value in several days which depends on the applied current. The anode was coated as soon as electrolysis began. Time required for the complete coverage increased with a decrease in the applied current. As the electrolysis progressed, there was a gradual loss (visual observation) in coating material; the greater the applied current, the greater the loss. Consequently, the appearance of the coatings on the anode after the termination of the experiment was intermediate of the two extremes - one compact and dense at 5 μA, and the other patchy and spotty at 400 μA. Unfortunately, we did not analyze the composition of the coating. Although the dissolution of gold on the wire surface was uniform, the rate of attack was severe at the tip and at the air-electrolyte-gold interface of the electrode. Consequently, several electrodes broke at the air-electrolyte-gold interface prior to 21 days.

We can not directly compare our data with Frankenthal and Siconolfi (5) data for the anodic corrosion of gold in NaCl because we have employed two electrode experiements and have no anode potential data. The major components (Table 1) of synthetic urine are chlorides and the concentration of $Cl^-$ ions is 0.14 M. A tentative explanation, however, could be drawn for the gold dissolution in urine at various applied currents by comparing the Frankenthal and Siconolfi data for the anodic corrosion of gold in 0.1M NaCl (5).

Table 3. Gold Dissolution in Synthetic Urine

| Current / μA | Cell Voltage / V After | | No. of Days | Weight Loss / mg | Percent Inhibition |
|---|---|---|---|---|---|
| | 1 Hr. | 7 Days | | | |
| 5 | 1.01 | 0.85 | 21 | 8.71 | 53.0 |
| 10 | 1.34 | 0.89 | 13 | 11.38 | 50.3 |
| 23 | 1.53 | 2.34 | 7 | 12.10 | 57.3 |
| 40 | 1.73 | No Data | <7 | 11.81 | 28.3 |
| 50 | 1.76 | 1.25 | 9 | 11.30 | 57.3 |
| 100 | 1.94 | 2.33 | 10 | 12.01 | 79.6 |
| 200 | 2.12 | 2.48 | 22 | 11.61 | 95.5 |
| 400 | 3.23 | 2.56 | 12 | 12.40 | 95.6 |




1. Time dependence of cell voltage for synthetic urine with (c and d) and without (a and b) TSB at 5 μA (b and d) and 400 μA (a and c) currents.

Currents up to 30 μA

$Au + 2Cl^- = AuCl_2^- + e^-$

$Au + organic(s) = Au.Organic(ad)$

Current range between 30-200 μA

$Au + 4Cl^- = AuCl_2^- + 3e^-$

$2Au + 3H_2O = Au_2O_3 + 6H^+ + 6e^-$

$Au + organic(s) = Au.Organic(ad)$

Currents above 200 μA

$Au + 4Cl^- = AuCl_4^- + 3e^-$

$2Cl^- = Cl_2 + 2e^-$

$Cl_2 + 2H_2O = HOCl + H_3O^+ + Cl^-$

$Au + organic(s) = Au.Organic(ad)$

Therefore, the observed gold inhibition in synthetic urine is due to the formation of passive films of oxide and hydroxide (5), organics (urea, creatinine etc. (Table 1 and ref. (6)).

The weight loss presented in Table 3 is the total weight loss at the termination of each experiment. The theoretical weight loss was calculated using a relationship 0.1764 It/n (Faradaic Law), where n = number of electrons, I = current in μA, and t = time of electrolysis in days. Per preceding discussion, the number of electrons used in the calculation were 1 and 3 for currents below and above 30 μA, respectively. From the calculated and the experimental weight losses, the percent dissolution efficiency of gold in urine was calculated. The percent inhibition given in Table 3 was calculated by subtracting the percent dissolution from 100. The percent inhibition of gold in synthetic urine was about 53 ± 4 up to 30 μA. After going through a minimum around 40 μA, the percent inhibition increased with an increase in current.

The data for the gold dissolution in synthetic urine with 5% TSB are given in Table 4. The cell voltage, in general, showed an initial raise followed by a decrease with time (Figure 1 (c and d) and Table 4). It is interesting to note that the cell voltages at *all applied currents in this solution* compared to the values in pure synthetic urine solution were initially lower, but changed to higher values as the electrolysis progressed (Figure 1 (c and d) and Tables 3 and 4). The general nature and the appearance of the coatings on the anode were similar to the one described above for the

**Table 4. Gold Dissolution in Synthetic Urine Containing 5% TSB**

| Current / μA | Cell Voltage / V After | | No. of Days | Weight Loss / mg | Percent Inhibition |
|---|---|---|---|---|---|
| | 1 Hr. | 7 Days | | | |
| 5 | 0.47 | 1.00 | 22 | 0.76* | >100 |
| 10 | 1.26 | 2.02 | 22 | 0.75 | 98.1 |
| 33 | 2.54 | 2.40 | 22 | 0.66 | 98.5 |
| 100 | 2.81 | 2.60 | 22 | 1.76 | 98.6 |
| 200 | 2.96 | 2.78 | 22 | 4.80 | 98.2 |
| 400 | 2.99 | 2.83 | 22 | 9.47 | 98.3 |

* weight gain

synthetic urine solution. However, the percent inhibition of gold dissolution calculated by the procedure described earlier was about 98.5 ± 0.5 (Table 4) and was greater in this solution compared to pure synthetic urine at all applied currents. Obviously, the adsorption of the nutrients in TSB (Table 2) further inhibits the gold dissolution in the synthetic urine containing 5% TSB.

Because infected urine (7) normally contains proteins, sugars and various unspecified substances, which act as nutrients for bacteria, the dissolution rate of gold under this condition would be considerably lower as is evident from this study. Not only does the low dissolution rate reduce the materials cost but it also regulates the release of gold ions at lower rate to kill the bacteria effectively over a three week period. In conclusion, the data helps to provide a basis for the design of catheter that may reduce or eliminate nosocomical and catheter - related sepsis.

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